IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Robert S. Whitehouse

Title: PHA ADHESIVE COMPOSITIONS

Appl. No.: 10/783,958

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Examiner: Saira Bano Haider

Art Unit: 1796

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DECLARATION OF ROBERT S. WHITEHOUSE UNDER 37 C.F.R. § 1.132

"The Whitehouse Declaration"

Dear Examiner Haider:

I, Robert S. Whitehouse, state and declare that:

1. I am a U.S. citizen residing at 159 Reed Street, Lexington, MA 02124.

2. I am a Senior Customer Applications Development Manager with Metabolix Corporation.

3. I received a Ph.D. in Polymer Chemistry / Adhesives from Wolverhampton University, UK, and have over 40 years of experience in the polymer and adhesives industry.

4. I am the inventor of the subject matter claimed in the above-referenced U.S. Patent Application No. 10/783,958.

5. In view of the comments of the Examiner in both the Office Action of March 12, 2010 and the interview which was held on June 29, 2010, the definition of adhesion appears to be critical to the understanding and differentiation of the claimed invention from that of the cited

references. To clarify, adhesion is the strength required to sever the interactions either between two substrates connected by an adhesive layer, or between an adhesive layer and a substrate. Adhesion may be shown by a variety of physical observations including surface tack, tack time, and the strength of a bond formed by applying the adhesive layer, or adhesive-coated substrate, to a second substrate under external application of pressure for a defined time interval. Generally, adhesive interactions between the adhesive layer and the substrate are attributable to short distance interactive forces, such as London forces, van der Waals forces, or hydrogen bonding. Stronger interactions, such as covalent or ionic bonds, typically only occur as a result of reactions, not mere adhesion. When an adhesive is applied to a surface the surface energy characteristics and viscoelastic flow of the adhesive contribute significantly to maximizing the interactive forces. Generally, *the adhesive should have a lower surface energy than the substrate*, and a lower viscosity promotes better wetting of the substrate.

An example demonstrating these interactive effects is the adhesion between a low surface energy substrate such as polyethylene, and a high surface energy substrate epoxide resin. Polyethylene has a surface energy of about 30 mJ/m^2, and epoxide resins have a surface energy of around $50mJ/m^2$. It is known that when an epoxide resin is applied to a polyethylene surface, it appears to wet the surface, however, this wetting is not at the molecular level and the epoxide resin can easily be removed. Conversely, if molten polyethylene is applied to a cured epoxide resin surface it readily wets the surface at the molecular level and the polyethylene is very difficult to remove from the cured epoxide.

6. PLA and PHA Systems As Described By Noda (U.S. Pat. No. US 6,808,795, published as 2002/0143136).

PLA has a surface energy of around 54 mJ/m^2, in comparison to PHAs which have surface energies that vary from about 50 mJ/m^2 for PHB (methyl side chain) and PHBV (ethyl side chain) to as low as 38 mJ/m^2 for PHO (polyhydroxyoctanaoate, pentyl side chain). PLA is typically amorphous (unless biaxially orientated) and has a Tg of +55°C. In comparison, PHAs

can vary from highly crystalline, *e.g.* PHB (Tg +7C, 65% crystallinity), to essentially amorphous, *e.g.* PHO (Tg -35C, 2-5% crystallinity).

In terms of surface energy, PLA has difficulty in wetting out onto a PHA substrate because of its higher surface energy. As a result, very poor adhesion is exhibited, as expected. Whereas, PHA, having a lower surface energy, would be expected to wet out the PLA surface and adhere. Unfortunately, because PHAs have to be applied at about 130-160°C and PLA has a Tg of +55C, the PLA substrate is rapidly distorting and melting as the PHA is deposited. Accordingly, the would-be PLA substrate, actually inhibits adhesion when the hot PHA is contacted to it.

The above discussion demonstrates the inoperability of a number of Noda's claims, if they are to be viewed as adhesives, and actually supports the express teachings of Noda with respect to "melt mixing" of the PLA and PHA polymers. *E.g. see* col. 11, line 65 – col. 13, line 67. In this section, Noda describes the use of various extrusion dies to being the melted polymers together to achieve melt mixing of the polymers. In other words, we believe that at the interface of the PLA and PHA the polymer strands intertwine to hold the different polymer materials together in a laminate or film. Without the intertwining of the polymer strands, there would not be adhesion as described above, and as that term is used in the present application.

7. Examples Of the Present Claims.

The present claims are directed to a unique range of PHA components having 4-HB (4-hydroxybutyrate) as a co-monomer with 3-HB (3-hydroxybutyrate), and which have a glass transition temperature range from about -30°C to about -5°C. Such PHAs are adhesives for a variety of surfaces, and they exhibit sufficient surface tack and tack times to be used effectively as adhesives.

Examples 13 through 22 demonstrate that a PHA based on poly(3-HB-co-4-HB) having Tg values from -30°C to -5°C, show good adhesion properties. The same examples also

illustrate that while a Tg from -30°C to -5°C is important in enhancing adhesive properties, molecular weight can also play a major role.

In contrast, Example 3 of the present application illustrates a PHA composition (poly(3HB)-co-8%-(3HV)) having a Tg of +2°C, which is outside that of the Tg recited in the claim window of -5°C to -30°C. The PHA composition of Example 3 is observed to exhibit *no* adhesion. It is known that adhesion of a substance may be promoted by the use of tackifiers. Yet, even when such tackifying resins (*e.g.* Foral 85 and Foral 105) are added to the PHA composition of Example 3, as set forth in Examples 4-7, no improvement in adhesion is observed. In fact the adhesion of Examples 4-7 is noted to be "negligible."

Further, when the Tg is significantly less than -30°C, adhesion is also negligible. This is shown by Example 8-9 of the present application, where a Tg of -60°C in a polycaprolactone polymer was too low, and the polymer exhibited negligible adhesion.

It is noteworthy to compare these results to the PHA polymer used by Noda, which is polyhydroxybutyrate-co-12% hydroxyhexanoate, and having a Tg of +2.8°C. In Example 1 of Noda, this polymer is listed as a preferred composition. However, based upon the above discussion, Noda's preferred polymer has a Tg that is outside the claimed range, and which is similar to that of the PHBV polymer in Example 3 of the present application. Accordingly, it is believed that the polyhydroxybutyrate-co-12% hydroxyhexanoate of Noda will not demonstrate good adhesive properties.

The examples in the present application demonstrate compositions that are different from those of Noda, and that have good adhesive properties. None of the PHAs described by Noda have the presently claimed Tg values, and none of the PHAs described by Noda are adhesives for the PLA materials to which they are melt-mixed.

8. I also conducted several additional experiments based upon the discussion that we had with Examiner Haider on June 9, 2010. To further illustrate the differences between the Noda

reference and the present invention, we tested the tack and open times of various PLA and PLA-PHA blends according to the procedures as described in the examples of USSN 10/783,958, the instant application. The PLA used was the commercially available Natureworks 5040. The PHAs used are as indicated in the table below. None of the PLA, or the PLA-PHA blends exhibited a tack or open time.

Material	Open Time (seconds)	Tack Time (minutes)
PLA	0	0
80/20 blend of PLA, poly-3-hydroxybutyrate and poly(3-hydroxybutyrate-co-11%-4-hydroxybutyrate)	0	0
20/80 blend of PLA, poly-3-hydroxybutyrate and poly(3-hydroxybutyrate-co-11%-4-hydroxybutyrate)	0	0

The above data supports the argument that (1) the PLA blends of Noda are not adhesives, and (2) if PLA were added to the adhesive compositions of the invention they would no longer be adhesives within the meaning of our invention.

9. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like are punishable by fine or imprisonment, or both (18 U.S.C. § 1001) and may jeopardize the validity of the above-referenced application or any patent issuing thereon.

Robert S. Whitehouse

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August 11th 2010

Date